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Spontaneous magnetization and Hall effect in superconductors with broken time-reversal symmetry

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Abstract. – Broken time reversal symmetry (BTRS) in d-wave superconductors is studied and is shown to yield current-carrying surface states. The corresponding spontaneous magnetization Φ is temperature independent near the critical temperature T_c for weak BTRS, in accord with recent data. For strong BTRS and thin films we expect a temperature-dependent Φ with a paramagnetic anomaly near T_c. The Hall conductance is found to vanish at zero wave vector q and finite frequency ω, however at finite q, ω it has an unusual structure.

Recent data on the high-T_c superconductor $YBa_2Cu_3O_x$ (YBCO) has supported the presence of broken time reversal symmetry (BTRS) [1–3]. A sensitive probe of BTRS are Andreev surface states. For a d-wave with time reversal symmetry bound states at zero energy are expected for a surface parallel to the nodes (a (110) surface in YBCO). When BTRS is present, by either a complex order parameter or by an external magnetic field, the bound states shift to a finite energy. Indeed tunneling data usually shows a zero-bias peak which splits in an applied field; the splitting is nonlinear in the magnetic field, indicating a proximity to a BTRS state [2,4]. In fact, in some samples tunnelling data shows a splitting even without an external field [1,2], consistent with BTRS; the splitting increases with increasing overdoping [2,5],

Further support for a spontaneous BTRS state are spontaneous magnetization data as observed in YBCO [3], setting in abruptly at T_c and being almost temperature (T) independent below T_c. The phenomenon has been attributed to either a $d_{x^2-y^2} + id_{xy}$ state or to the formation of π junctions. No microscopic reason was given, however, for the spontaneous magnetization being independent of both T and of film thickness [3].

It has been shown theoretically that BTRS can occur locally in a $d_{x^2-y^2}$ superconductor near certain surfaces [6–8] leading to surface currents. The onset of such BTRS is expected to be below T_c and therefore does not correspond to the spontaneous magnetization data [3]. We note that in response to an external magnetic field the surface states are paramagnetic and compete with Meissner currents. This effect has been proposed to account for a minimum in the magnetic penetration length [9]. In fact, it was proposed that this paramagnetic effect leads to spontaneous currents and BTRS in a pure $d_{x^2-y^2}$ state [10, 11]. The onset of this BTRS is much below T_c [11] and therefore does not correspond to the data [3].

Of further theoretical interest is the relation of the BTRS state to quantum Hall systems with a variety of Hall effects [12–15]. In particular a finite charge Hall conductance has been suggested [12], though this has been questioned [14].

In the present work we assume that BTRS is a bulk property, *i.e.* both components of an order parameter $d_{x^2-y^2} + id_{xy}$ set in at T_c; this is a plausible scenario which can possibly account for the data. In previous works [6–8] surface states appear due to a surface-induced id_{xy} component. We find, however, the less anticipated result that the *bulk* state $d_{x^2-y^2} + id_{xy}$ leads to surface states with finite surface current densities. The latter situation was found in the bulk p-wave state [16] and for the total current was inferred from topological considerations [17]. We then evaluate the spontaneous magnetization and show that it is dominated by (100) surfaces; for thin films it increases with the ratio Δ'/Δ (Δ and Δ' are the amplitudes of $d_{x^2-y^2}$ and d_{xy}, respectively) while for thick films it has a maximum at $\lambda/\xi' \approx 1$, where ξ, ξ' are the coherence lengths associated with Δ, Δ', respectively (ξ' = Fermi velocity$/\Delta'$, similarly for ξ) and λ is the magnetic penetration length. The maximum for YBCO is at $\Delta'/\Delta \approx \xi/\lambda \approx 0.01$. Throughout we assume an exterme type-II superconductor, *i.e.* $\xi \ll \lambda$, while ξ'/λ is arbitrary. We show that for weak BTRS, $\lambda/\xi' < 1$, the spontaneous magnetization is T- and thickness-independent, while for strong BTRS thickness- and T-dependence may occur, as well as a transition to a paramagnetic state close to T_c. For the sample of ref. [3] we estimate $\Delta'/\Delta \approx 10^{-4}$, *i.e.* weak BTRS. We also derive the effective action and identify the Hall coefficient which has an unusual wave vector and frequency dependence.

First we demonstrate the existence of surface states. Consider a $d_{x^2-y^2} + id_{xy}$ state where the order parameter is

$$\Delta(\hat{p}_x, \hat{p}_y) = \Delta'\hat{p}_x\hat{p}_y/k_F^2 + i\Delta(\hat{p}_x^2 - \hat{p}_y^2)/k_F^2\,, \tag{1}$$

where $\hat{\boldsymbol{p}} = -i\hbar\boldsymbol{\nabla}$ is the momentum operator and k_F is the Fermi momentum. We consider a vacuum-superconductor boundary at $x = 0$, and assume for now that Δ, Δ' are constants at $x > 0$ and vanish at $x < 0$. For $\Delta \gg \Delta'$ this corresponds to a (100) surface; to describe a (110) surface Δ and Δ' need to be interchanged. The electron-hole wave functions $u(x, k_y)\exp[ik_y y]$, $v(x, k_y)\exp[ik_y y]$ with mass m satisfy the Bogoliubov-de Gennes equations

$$\begin{aligned}(2m)^{-1}(-k_F^2 - \mathrm{d}^2/\mathrm{d}x^2)u(x, k_y) + \Delta(\hat{p}_x, k_y)v(x, k_y) &= \epsilon u(x, k_y)\,,\\ \Delta^*(\hat{p}_x, k_y)u(x, k_y) + (2m)^{-1}(k_F^2 + \mathrm{d}^2/\mathrm{d}x^2)v(x, k_y) &= \epsilon v(x, k_y)\,.\end{aligned} \tag{2}$$

The decaying eigenfunctions have the form $\sim \exp[\pm ikx - x|k_y|/\xi' k_F]$, with $k = |k|$ and $\xi' = k_F/m\Delta'$. Specular reflection at the surface requires a superposition of $\pm k$ states which vanish at the surface. This yields the eigenvalue equation

$$\frac{i\epsilon + \sqrt{|\Delta(+k, k_y)|^2 - \epsilon^2}}{-i\epsilon + \sqrt{|\Delta(-k, k_y)|^2 - \epsilon^2}} = -\frac{\Delta(+k, k_y)}{\Delta(-k, k_y)}\,. \tag{3}$$

Its solutions are $\epsilon = -\mathrm{sign}(k_y)\Delta(k^2 - k_y^2)/k_F^2$. In terms of the incidence angle ζ, $k_y = k_F \sin\zeta$, $k = k_F\cos\zeta$, the allowed positive eigenvalues are $\epsilon = \Delta\cos(2\zeta)$ for $-\pi/4 < \zeta < 0$ and $\epsilon = -\Delta\cos(2\zeta)$ for $\pi/4 < \zeta < \pi/2$ (see inset of fig. 1). We note that self-consistency would imply that $\Delta' = 0$ at $x = 0$ [8]; the eigenfunctions would then be $\sim \exp[-\int_0^x \Delta'(x')\mathrm{d}x'|\sin\zeta|/v_F]$, resulting in a qualitatively similar dependence on ξ'. The dominant order parameter Δ is finite at the (100) boundary [8], hence we expect our results to be quantitatively correct.

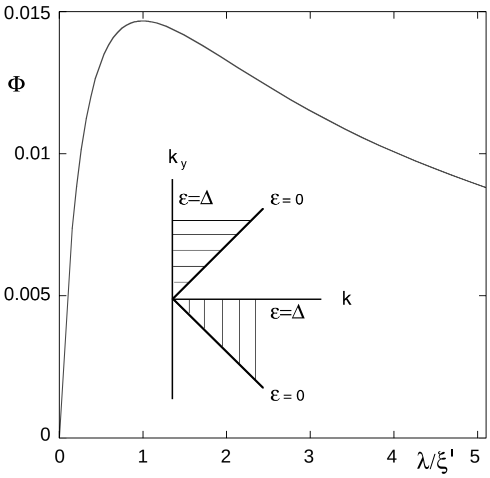

Fig. 1 – Spontaneous flux for a (100) boundary in thick films ($\xi' < \bar{d}$) in units of $2\phi_0 L_y \lambda\Delta/\pi\lambda_0^2 T_c$. The hatched areas in the inset show the allowed directions (k, k_y) of bound states. Their spectra span the range $\epsilon = 0$ (diagonal lines) up to $\epsilon = \Delta$ as shown.

In the presence of a vector potential $A_y(x)$ the spectrum ϵ is Doppler-shifted by $-(e/mc)k_y A_y$. For the expectation value of the current density in the y-direction and the charge density, we obtain

$$\begin{aligned} \frac{4\pi}{c} j_{\rm edge}(x) &= \frac{4\phi_0}{\pi\xi'\lambda_0^2}\int_0^{\pi/2} {\rm d}\zeta \cos\zeta \sin^2\zeta e^{-2x\sin\zeta/\xi'} \tanh\left(\frac{\Delta\cos 2\zeta + (e/c)v_{\rm F}\sin\zeta A_y(x)}{2T}\right), \\ n_{\rm edge}(x) &= \frac{ek_{\rm F}}{\pi {\rm d}\xi'}\int_0^{\pi/2} {\rm d}\zeta \cos\zeta \sin\zeta e^{-2x\sin\zeta/\xi'}, \end{aligned} \tag{4}$$

where $\lambda_0 = (mc^2 d/2k_{\rm F}^2 e^2)^{1/2} = \lambda(T=0)$ and d is the spacing between layers, converting the current per layer of the states in eq. (2) to a current density. Note that for either $\Delta = 0$ or $\Delta' = 0$ all angles ζ are allowed in the solution of eq. (3) and then the current density vanishes. This demonstrates then that BTRS leads to current-carrying surface states. We note also that the integrated current $\int_0^\infty j_{\rm edge}(x){\rm d}x$ vanishes, unlike the p-wave case [16].

The response to $j_{\rm edge}(x)$ includes London terms as well as a BTRS-induced Chern-Simon term. As shown below, the Chern-Simon term is weaker at $T \to T_{\rm c}$ so that London's equation with $j_{\rm edge}(x)$ as a source term is valid,

$$-\nabla^2 A_y(x) = [-(1/\lambda^2)A_y(x) + (4\pi/c)j_{\rm edge}(x)]\theta(x), \tag{5}$$

where $\theta(x)$ is a step function. We consider first solutions where the Doppler shift $\sim A_y$ on the right-hand side of eq. (4) is neglected. This is solved by the Greens' function $G(x,x') = -(\lambda/2)[\exp[-|x-x'|/\lambda] + \exp[-|x+x'|/\lambda]]$ resulting at $T \to T_{\rm c}$ in the solution

$$A_y(0) = (2\phi_0\lambda\Delta/\pi\lambda_0^2 T_{\rm c})\int_0^{\pi/2} {\rm d}\zeta \cos\zeta \sin^2\zeta \cos 2\zeta (2\sin\zeta + \xi'/\lambda)^{-1}. \tag{6}$$

The total spontaneous flux is $\Phi = A_y(0)L_y$, where L_y is the length of the boundary. The ratio $\tilde{\Phi} = -\Phi/(2\phi_0 L_y \lambda\Delta/\pi\lambda_0^2 T_{\rm c})$ is shown in fig. 1. It varies between $\xi'/12\lambda$ at $\lambda \gg \xi'$ to

$\lambda/15\xi'$ at $\lambda \ll \xi'$ with a maximum of 0.014 at $\lambda \approx \xi'$. For a (110) surface $\tilde{\Phi} = \xi\Delta'/12\lambda\Delta$, much smaller than for a (100) surface. We note that Φ is T-independent up to $T_{\rm c}$ since the product $\lambda\Delta$ is finite at $T \to 0$, consistent with the spontaneous magnetization data [3]; more details on the data follow in the discussion.

The above refers to thick films with thickness $\bar{d} > \xi'$. For thin films the right-hand side of eq. (5) is multiplied by $\bar{d}\delta(z)$. For $A_y(x) \equiv A_y(x, z = 0)$ this yields

$$A_y(x) - A_y(0) = \int_0^\infty \mathrm{d}x' \ln|(x - x')/x'|[-(1/\lambda^2)A_y(x') + (4\pi/c)j_{\rm edge}(x')]\bar{d}\,, \qquad (7)$$

which implies a slow (nonexponential) decay of $A_y(x)$. To avoid divergence of $\mathrm{d}A_y(x)/\mathrm{d}x|_{x=0}$, the relation $A_y(0) = \lambda^2(4\pi/c)j_{\rm edge}(0)$ must hold. This, interestingly, yields for Φ the previous result of the $\lambda \ll \xi'$ case, *i.e.* $\tilde{\Phi} = \lambda/15\xi'$; in fig. 1 this is the tangent line to the thick film curve at the origin. Hence we can define two regimes: Weak BTRS with $\lambda/\xi' < 1$ where the spontaneous flux is T- and $\bar{d}$-independent, and strong BTRS with $\lambda/\xi' > 1$ where film thickness matters. In the latter case the T-dependence is induced as $\xi' < \bar{d}$ changes to the thin-film case $\xi' > \bar{d}$ as $T \to T_{\rm c}$.

We now reconsider the effect of the Doppler shift. Near $T_{\rm c}$ the effect is linear in A_y and can be incorporated as an effective $\lambda_{\rm eff}$ replacing λ in the results above, where

$$\frac{1}{\lambda_{\rm eff}^2} \approx \frac{1}{\lambda^2} - \frac{\xi_0}{\lambda_0^2 \max(\xi', \lambda)}\,. \qquad (8)$$

Hence at a temperature $T'_{\rm c}$ $\lambda_{\rm eff}$ changes sign, where $(T_{\rm c} - T'_{\rm c})/T_{\rm c} \approx [\xi/\max(\xi', \lambda)]^2$, *i.e.* less then 10^{-4} for YBCO. The effect of the Doppler shift was previously considered only at low temperatures where on the (110) surface paramagnetism and spontaneous surface currents appear even when $\Delta' = 0$ at $\sim (\xi_0/\lambda_0)T_{\rm c} \ll T_{\rm c}$ [10,11]. For thin films, $\max(\xi', \lambda)$ is replaced by ξ' and $(T_{\rm c} - T'_{\rm c})/T_{\rm c} \approx (\xi/\xi')^2$ also for strong BTRS where $T_{\rm c} - T'_{\rm c}$ is enhanced. Hence a transition from paramagnetic ($T > T'_{\rm c}$) to diamagnetic response ($T < T'_{\rm c}$) can be observed at $T'_{\rm c}$ and the magnetization $\tilde{\Phi} = (\lambda/15\xi')(\lambda_{\rm eff}/\lambda)^2$ is peaked and changes sign at $T'_{\rm c}$. The effect is more pronounced for the (110) surface, however, the condition for a thin film $\bar{d} < \xi$ is more difficult to achieve.

Next we consider the effective action of a bulk $d_{x^2-y^2} + id_{xy}$ superconductor. In terms of the Nambu spinors $\psi^\dagger(\boldsymbol{r}) = [u^*(\boldsymbol{r}), u^*(\boldsymbol{r})]$, the superconducting phase $\theta(\boldsymbol{r})$ and Pauli matrices τ_i, the transformation $\psi(\boldsymbol{r}) \to \exp[i\tau_3\theta(\boldsymbol{r})/2]\psi(\boldsymbol{r})$ yields the off-diagonal Hamiltonian $\int \mathrm{d}^2r\psi^\dagger(\boldsymbol{r})h_\Delta\psi(\boldsymbol{r})$, where

$$h_\Delta = -[\Delta(-\partial_x^2 + \partial_y^2)\tau_1 + \Delta'\partial_x\partial_y\tau_2]/k_{\rm F}^2 \qquad (9)$$

and we neglect terms with $\nabla\theta \ll k_{\rm F}$. The action in the presence of electromagnetic potentials $\boldsymbol{A}, \varphi$ is then

$$\begin{aligned} S &= \int \mathrm{d}^2r\mathrm{d}t\psi^\dagger(i\partial_t - \tau_3\epsilon(\hat{p}) - h_\Delta - \Sigma)\psi\,, \\ \Sigma &= \tau_3(a_0 + \boldsymbol{a}^2/2m) + \boldsymbol{a}\cdot\boldsymbol{p}/m - i\boldsymbol{\nabla}\cdot\boldsymbol{a}/2m\,, \end{aligned} \qquad (10)$$

where $\epsilon(\hat{p}) = (\hat{p}^2 - k_{\rm F}^2)/2m$ and we introduce the gauge-invariant potentials $\boldsymbol{a} = \frac{1}{2}\boldsymbol{\nabla}\theta - e\boldsymbol{A}$ and $a_0 = \frac{1}{2}\frac{\partial}{\partial t}\theta - e\varphi$. Expansion to 2nd order in $\boldsymbol{a}, a_0$ leads to the effective action

$$S_{\rm eff} = \int \frac{\mathrm{d}^2q\mathrm{d}\omega}{(2\pi)^3} P_{\mu\nu}(\boldsymbol{q}, \omega)a_\mu(\boldsymbol{q}, \omega))a_\nu(-\boldsymbol{q}, \omega))\,. \qquad (11)$$

At $T = 0$ and $\boldsymbol{q}, \omega \to 0$ we obtain $P_{00} = N_0$ (density of states which is $N_0 = m/2\pi$ in two dimensions), $P_{ij} = -N_0 c_s^2$, where $c_s^2 = v_{\mathrm{F}}/\sqrt{2}$, while $P_{0j}(q) = i\mathrm{sign}(\Delta\Delta')\epsilon_{0ij}q_i/(4\pi)$ and ϵ_{0ij} is the antisymmetric unit tensor. The latter term reflects BTRS and is derived for $\Delta' \ll \Delta$.

Integrating out the phase θ, we obtain the effective action in terms of the electromagnetic potentials $\boldsymbol{A}, \varphi$

$$S_{\mathrm{eff}}\{\boldsymbol{A}, \varphi\} = e^2 \int \frac{\mathrm{d}^2 q \mathrm{d}\omega}{(2\pi)^3} \left\{ \frac{c_s^2 \boldsymbol{q}^2}{c_s^2 \boldsymbol{q}^2 - \omega^2} \left[P_{00} |\varphi(\boldsymbol{q}, \omega)|^2 - \frac{i}{4\pi} \epsilon_{0ij} q_i \varphi(\boldsymbol{q}, \omega) A_j(-\boldsymbol{q}, -\omega) + \right.\right.$$
$$\left.\left. + O(\omega^2 |\boldsymbol{A}|^2) \right] - P_{00} \left(\frac{c_s}{c} \right)^2 |\boldsymbol{A}(\boldsymbol{q}, \omega)|^2 \right\}. \tag{12}$$

The total electromagnetic action includes also the Maxwell field part $S_{\mathrm{M}} = \int \mathrm{d}^2 r \mathrm{d}t (\vec{E}^2 - \vec{H}^2)/8\pi$. *E.g.*, for $\omega \neq 0$, $q \to 0$ the propagator for φ yields the plasmon mode at $\omega_p = c/\lambda_0$. The Hall current J_y is identified by a functional derivative with respect to A_x leading to the Hall coefficient

$$\sigma_{xy}(\boldsymbol{q}, \omega) = \mathrm{sign}(\Delta\Delta') \frac{e^2}{4\pi\hbar} \frac{c_s^2 q^2}{c_s^2 q^2 - \omega^2}. \tag{13}$$

Transport is defined by taking the $q \to 0$ limit first, *i.e.* $\sigma_{xy} = 0$. Hence the conventional Hall coefficient vanishes, as expected from Galilean invariance [14]. A limit in which $\omega \to 0$ is taken first yields a quantized "static" conductance $e^2/2h$ which was argued to correspond to $\sigma_{xy} \neq 0$ in the presence of a boundary [12]. In the absence of an external magnetic field, and given a spontaneous magnetization decaying in the bulk (as confirmed below), Ampère's law yields zero total current, hence $\sigma_{xy} = 0$; this is valid also with a boundary and external electric field. It is intriguing, however, that $\sigma_{xy}(\boldsymbol{q}, \omega)$ has a nontrivial structure and space-resolved measurement of a Hall current could then probe the full equation (13). We note that a result similar to eq. (13) was obtained for superfluid ^{3}He [15].

We proceed to derive the effective action in the presence of a boundary and at finite T. Special care is needed for the Chern-Simon coefficient P_{0j} which is now nonlocal due to the specular reflection at the boundary. After integrating out θ, we obtain the form (considering only the $\omega = 0$ term)

$$S_b\{\boldsymbol{A}, \varphi\} = e^2 \int \mathrm{d}r \left[P_{00} \left(\varphi^2(\boldsymbol{r}) - \left(\frac{c_s}{c} \right)^2 \boldsymbol{A}^2(\boldsymbol{r}) \right) + b_1(\boldsymbol{r}) \varphi(\boldsymbol{r}) A_y(\boldsymbol{r}) + b_2(\boldsymbol{r}) \varphi(\boldsymbol{r}) \frac{\partial A_y(\boldsymbol{r})}{\partial x} \right]. \tag{14}$$

Variation of $S + S_{\mathrm{M}}$ leads to a generalization of eq. (5) in which the equations for $\boldsymbol{A}$ and φ are coupled,

$$\left(\frac{1}{\lambda_{\mathrm{D}}^2} - \frac{\partial^2}{\partial x^2} \right) \varphi - \frac{4\pi e^2}{c\hbar} b_2 \frac{\partial A_y}{\partial x} - \frac{4\pi e^2}{c\hbar} b_1 A_y = 4\pi e n_{\mathrm{edge}}(x), \tag{15}$$

$$\left(\frac{1}{\lambda^2} - \frac{\partial^2}{\partial x^2} \right) A_y - \frac{4\pi e^2}{c\hbar} b_2 \frac{\partial \varphi}{\partial x} + \frac{4\pi e^2}{c\hbar} (b_1 - \partial_x b_2) \varphi = -\frac{4\pi}{c} j_{\mathrm{edge}}(x), \tag{16}$$

where $\lambda_{\mathrm{D}} = (8\pi e^2 N_0)^{-1/2}$ is the Debye screening length. For $T \to T_{\mathrm{c}}$ we obtain $\lambda \approx \lambda_0 (1 - T/T_{\mathrm{c}})^{-1/2}$, $b_1(\boldsymbol{r}) = 0.11(\Delta\Delta'/T_{\mathrm{c}}^2) \frac{\mathrm{d}}{\mathrm{d}x} \ln[\Delta\Delta']/2hcd$ and $b_2(\boldsymbol{r}) = 0.21(\Delta\Delta'/T_{\mathrm{c}}^2)/2hcd$.

An external electric field leads to $\varphi \sim \exp[-x/\lambda_{\mathrm{D}}]$, hence the magnetization remains localized even in presence of such a field and the space integration of eq. (16) (*i.e.* Ampère's law) yields a zero total current, *i.e.* $\sigma_{xy} = 0$. The Chern-Simon term, however, affects the spontaneous magnetization, leading to an additional flux $\sim (\Delta'/\Delta)(1 - T/T_{\mathrm{c}})$ which vanishes at $T \to T_{\mathrm{c}}$.

We consider now in more detail the experimental data on the spontaneous magnetization [3]. The data shows that for a YBCO disc with a perimeter of $L_y \approx 2\,\mathrm{cm}$ the spontaneous magnetization is temperature independent in the range 80–89 K and is also thickness-independent in the range 30–300 nm with a value of $\approx 37\phi_0$. Taking $\lambda\Delta \approx \lambda_0\Delta_0$, their $T = 0$ value, and typical YBCO parameters we find $\tilde{\Phi} \approx 10^{-3}$. Figure 1 implies that the limit $\xi' > \lambda$ applies, *i.e.* weak BTRS; hence for either thick or thin films we estimate $\lambda/\xi' \approx 10^{-2}$ or $\Delta'/\Delta \approx 10^{-4}$. We propose therefore that increasing the ratio Δ'/Δ, *e.g.* by using overdoped YBCO [2], one can enhance the spontaneous magnetization up to a maximum of $\approx 10^3\phi_0$ when $\Delta'/\Delta \approx 0.01$ within the thick-film regime.

For strong BTRS, $\lambda/\xi' > 1$, the film thickness matters, *i.e.* we expect a temperature dependence due to the crossover from thick- to thin-film regimes at $\bar{d} \approx \xi'$ as $T \to T_c$. For thin films ($\bar{d} < \xi' < \lambda$) we obtain $\tilde{\Phi} = \lambda/12\xi'$, *i.e.* for YBCO the total flux can reach $10^5\Delta'/\Delta\phi_0$ per cm of boundary, much higher than thick-film values. The situation of a strong BTRS with thin films is interesting also as being the most likely one to show the paramagnetic anomaly at $T'_c \approx T_c[1 - (\xi/\xi')^2]$.

In conclusion, we have shown that the surface states of a $d_{x^2-y^2} + id_{xy}$ superconductor lead to a spontaneous magnetization which is T-independent and thickness-independent for weak BTRS, $\lambda/\xi' < 1$, in accord with the data [3]. For strong BTRS, $\lambda/\xi' > 1$, as expected in overdoped YBCO [2], a crossover from thick to thin film behavior can lead to T- and thickness-dependence, as well as to an observable paramagnetic anomaly near T_c. We also find that the Hall conductance has an unusual $\sigma_{xy}(\boldsymbol{q}, \omega)$ dependence, though its conventional transport value vanishes.

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